

Recommended Collision Integrals for Transport Property Computations, Part 1: Air Species

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A review of the best-available data for calculating a complete set of binary collision integral data for the computation of the mixture transport properties (viscosity, thermal conductivity, and ordinary and thermal diffusion) of 13-species weakly ionized air is presented. Although the fidelity of the data varies, all collision integrals presented herein, except for electron-neutral interactions, are estimated to be accurate to within 25% over the temperature range of interest (300–15,000 K) for reentry and laboratory plasmas. In addition, most of the dominant atom–atom and atom–ion interactions for dissociated weakly ionized air were derived from ab initio methods that are estimated to be accurate to within 10%. The accuracy and valid temperature range for electron-neutral interactions vary because of scarcity of the required cross-sectional data.

Nomenclature

| | | |
|----------------|---|--|
| B^*, C^* | = | nondimensional collision integral ratios |
| E | = | interaction energy, eV |
| e | = | electron charge (4.803×10^{-10} esu) |
| h | = | Planck's constant (6.626×10^{-27} erg · s) |
| k | = | Boltzmann constant (1.3806×10^{-16} erg/K) |
| n_e | = | number density of electrons, cm^{-3} |
| r | = | separation distance, cm |
| s | = | quantum spin number |
| T | = | temperature, K |
| T^* | = | reduced temperature |
| θ | = | scattering angle, deg |
| Λ^* | = | de Boer parameter |
| λ_D | = | Debye length, cm |
| μ | = | reduced mass, g |
| σ | = | cross section, cm^2 |
| φ | = | potential energy |
| $\Omega^{1.1}$ | = | diffusion collision integral, \AA^2 |
| $\Omega^{2.2}$ | = | viscosity collision integral, \AA^2 |

Subscripts

| | | |
|-------|---|--------------------|
| class | = | classical |
| ex | = | exchange |
| m | = | momentum transfer |
| qm | = | quantum mechanical |
| v | = | viscosity |

Introduction

ACCURATE modeling of the transport properties of weakly ionized air is important in several fields, including the aerothermodynamics of reentering spacecraft and the study of laboratory plasmas. To compute the transport properties of a dilute gas mixture, collision data are needed as a function of temperature for all binary interactions that occur in the gas. For weakly ionized air, the

mixture can be described with 13 species: N_2 , N_2^+ , O_2 , O_2^+ , NO , NO^+ , N , N^+ , O , O^+ , Ar , Ar^+ , and e^- , which leads to a total of 91 possible binary interactions.

According to Chapman–Enskog theory, as described in Ref. 1, the transport properties of a gas mixture can be computed by solving the Boltzmann equation using a Sonine polynomial expansion. Because of the rapid convergence of this expansion, the coefficients of viscosity, thermal conductivity, and ordinary diffusion can typically be accurately represented using only the first term. (See Refs. 2 and 3 for a discussion of the accuracy requirements for viscosity and thermal conductivity coefficients.) The resulting expressions, given in Ref. 1, require knowledge of only three binary interaction parameters: the diffusion collision integral $\Omega^{1.1}$, the viscosity collision integral $\Omega^{2.2}$, and the dimensionless collision integral ratio $B^* = (5\Omega^{1.2} - 4\Omega^{1.3})/\Omega^{1.1}$. One additional dimensionless quantity, $C^* = \Omega^{1.2}/\Omega^{1.1}$, is necessary to evaluate the second term of the expansion as required to compute thermal diffusion coefficients. Higher-order terms of the Sonine expansion, which may be required for highly accurate computations of the electron thermal conductivity in a partially ionized gas, introduce additional collision integral ratios.

Several reviews of collision integrals for weakly ionized air have been published in the past two decades.^{4–7} Notably, the data of Gupta et al.⁴ are still widely used; however, they are based primarily on analytical potentials developed during the 1960s that have largely become obsolete. Murphy and Arundell⁵ and Murphy⁶ published comprehensive reviews of collision integral data for air species, collecting data from multiple sources of varying fidelity. Most recently, Capitelli et al.⁷ computed collision integrals from analytic potentials for all air interactions over a temperature range of 50–50,000 K. However, the focus of this work was on very high temperatures,⁷ and therefore the methods used for interactions involving molecules often relied on exponential repulsion (neutral–neutral) or polarization (ion–neutral) potential models. In addition, none of these reviews account for the recent ab initio computations of several important atom–molecule,⁸ molecule–molecule,⁹ and ion–neutral^{10,11} air interactions.

The purpose of this work is to collect in a single place the best available collision integral data for weakly ionized air species at temperatures of interest for Earth entry and laboratory air plasmas (~300–15,000 K). The data presented herein can be readily fitted to an appropriate expression^{4,12} for use in existing computational fluid dynamics codes. Data are presented for species in the ground electronic state only, which is appropriate for the conditions of interest in most reentry problems. The effect of electronic excitation on transport collision integrals has been computed for some low-lying states of oxygen and nitrogen atoms^{13,14} as well as for hydrogen atoms.¹⁵ In addition, Capitelli et al.¹⁶ have looked at the effect of electronically excited states on local thermodynamic equilibrium hydrogen

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plasmas. The hydrogen results of Celiberto et al.¹⁵ are presented in a form that can be readily extended to other species if required.

Results

Neutral–Neutral Interactions

For the 21 possible interactions between two neutral atomic or molecular species, preference is given to recent ab initio calculations from quantum-mechanically derived potential energy surfaces that include extensive benchmark comparisons to previous theoretical and experimental results.^{8,9,17} This set includes the nitrogen–oxygen atom–atom interactions (N–N, N–O, O–O) and several of the most important atom–molecule and molecule–molecule interactions, including N₂–N₂, N₂–N, and O₂–O. The estimated accuracy of these computations is approximately 5% for atom–atom interactions and 10% for atom–molecule and molecule–molecule interactions over the temperature range 300–15,000 K.

When such ab initio calculations do not exist, alternate sources are used. In 1975, Cubley and Mason¹⁸ conducted beam-scattering experiments to measure the high-energy spherically averaged scattering potential of all atom–molecule and molecule–molecule interactions in nonionized air. Data were obtained for scattering beams of Ar atoms by Ar, N₂, and O₂. Atom–atom, atom–molecule, and molecule–molecule potentials for all neutral interactions were then obtained from these results via semiempirical combination rules. The resulting data were fit to exponential repulsion potentials, for which tabular expressions for the transport collision integrals have been published.¹⁹ These results should be reasonably accurate at elevated temperatures (>2000 K), because the exponential repulsion potential has the correct asymptotic form at high energies. The stated accuracy of the resulting collision integrals was 5% over the temperature range 300–15,000 K (Ref. 18). However, the use of semiempirical combination rules to construct potentials from simpler “building blocks” does not account for the details of the atom–molecule and molecule–molecule potential energy surfaces. Therefore, a better estimate of the accuracy of the Cubley and Mason data can be obtained by comparison to recent ab initio calculations of Stallcop et al.^{8,9} where the two data sets overlap. Figure 1 shows the results of this comparison for the viscosity collision integral ($\Omega^{2,2}$) of the N₂–N₂, N₂–N, and O₂–O interactions. The Cubley and Mason results are within 10% of the ab initio data below about 2500 K, but larger discrepancies are seen at higher temperatures for N₂–N₂ and N₂–N. Results for the diffusion collision integral (not shown), are nearly identical. Based on these results a more realistic accuracy estimate for the Cubley and Mason data is approximately 25% for atom–molecule and molecule–molecule interactions. The experimental data of Cubley and Mason¹⁸ have been used extensively to validate later theoretical work and remain the best source of data for several minor air atom–molecule interactions that have not been studied extensively since that time, including O₂–N, NO–N, NO–O, and Ar–N.

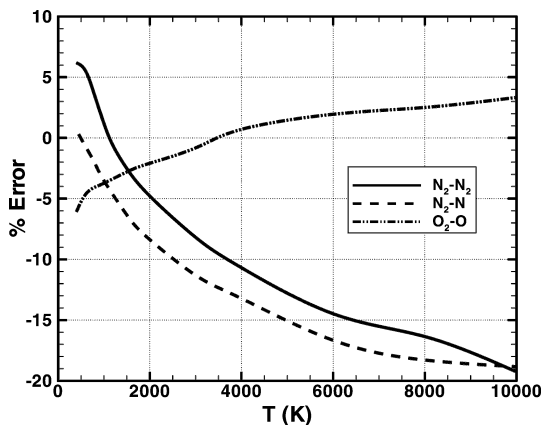


Fig. 1 Relative difference of the collision integral $\Omega^{2,2}$ for the N₂–N₂, N₂–N, and O₂–O interactions computed from the Cubley and Mason¹⁸ potential data as compared to the recent ab initio calculations of Stallcop et al.^{8,9}

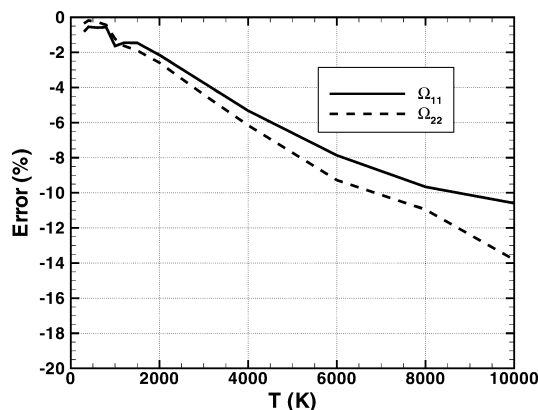


Fig. 2 Relative difference of the collision integrals $\Omega^{1,1}$ and $\Omega^{2,2}$ for the N₂–N₂ interaction computed by using the Bzowski et al.²⁶ universal collision integral method as compared to the recent ab initio calculations of Stallcop et al.⁹

For the N₂–O₂, N₂–O, Ar–N₂, Ar–O₂, and Ar–O interactions, we rely on the compilations of Murphy and Arundell⁵ and Murphy.⁶ The collision integrals for each of these interactions were integrated from high-fidelity potentials^{20–23} that account for the experimental data from Cubley and Mason¹⁸ at high temperature and include low-temperature corrections to account for dispersive and other long-range forces. These data have an estimated accuracy of about 20% over the temperature range 300–15,000 K.^{5,6} Collision integrals for Ar–Ar interactions were also taken from Murphy and Arundell⁵ and were based on a quantum-mechanical ab initio interaction potential from Aziz and Slaman.²⁴ The species viscosity for Argon computed using these values agrees with experimental values reported by Bich et al.²⁵ to within 5% up to 5000 K.

Finally, Bzowski et al.²⁶ developed a universal collision-integral concept and applied it to binary mixtures of polyatomic and noble gases. The binary interaction constants for each pair were chosen to best match the available experimental and theoretical data for the interaction. This method was shown to be in good agreement (within $\pm 15\%$) with existing pure-species experimental data,²⁶ although the error will be somewhat larger for interactions between different species because of the somewhat empirical nature of the mixing rules employed. Figure 2 shows the relative error of the collision integrals $\Omega^{1,1}$ and $\Omega^{2,2}$ for the N₂–N₂ interaction computed by using the Bzowski et al. method as compared to the recent ab initio calculations of Stallcop et al.⁹ The two agree to within 15% over the temperature range 300–10,000 K. The Bzowski et al. universal collision integrals are used here for the binary interactions O₂–O₂, NO–O₂, NO–N₂, NO–NO, and Ar–NO.

Tables 1 and 2 show the recommended values for the collision integrals $\Omega^{1,1}$ and $\Omega^{2,2}$ for all 21 neutral–neutral interactions that occur in 13-species air, as well as an estimate of the accuracy (Acc.) of these data over the temperature range 300–15,000 K. The final column of the tables lists the references used to determine their values. Values are not given for all interactions at all temperatures; this is because for some of the interactions the data were published at discrete temperatures, and the temperatures reported differ among the references. The dimensionless collision-integral ratios B^* and C^* are only weak functions of temperature, and a constant value of $B^* = 1.15$ and $C^* = 0.92$ can be used to represent all neutral–neutral interactions to within 10% accuracy (Table 3), which is sufficient for reentry aeroheating applications.

Ion–Neutral Interactions

Recommended collision integrals for all 36 ion–neutral interactions in 13-species air were recently presented in tabular form in Refs. 10 and 11 and are not repeated here. Data for most of these interactions were obtained by assuming a modified Tang–Toennies interaction potential,²⁷ in which the long-range form is governed by polarization and dispersion forces and the short-range form is effectively an exponential repulsion. The resulting collision integrals were found to agree to within 20% over the

Table 1 Diffusion collision integral $\Omega^{1,1}$ (\AA^2) as a function of temperature for neutral–neutral and electron–neutral interactions in air

| Interaction | T, K | | | | | | | | | | | | Acc.,% | Ref. |
|--------------------------------|-------|-------|-------|------|------|------|------|------|------|--------|--------|--------|--------|--------|
| | 300 | 500 | 600 | 1000 | 2000 | 4000 | 5000 | 6000 | 8000 | 10,000 | 15,000 | 20,000 | | |
| N ₂ –N ₂ | 12.23 | — | 10.60 | 9.79 | 8.60 | 7.49 | — | 6.87 | 6.43 | 6.06 | — | — | 10 | 9 |
| N ₂ –O ₂ | 10.16 | — | — | 7.39 | 6.42 | 5.59 | 5.35 | — | — | 4.60 | 4.20 | — | 20 | 5 |
| N ₂ –NO | 11.88 | 10.61 | 10.24 | 9.35 | 8.12 | 6.82 | 6.43 | 6.12 | 5.66 | 5.31 | 4.71 | — | 25 | 24 |
| N ₂ –N | 10.10 | — | 8.57 | 7.70 | 6.65 | 5.65 | — | 5.05 | 4.61 | 4.25 | — | — | 10 | 8 |
| N ₂ –O | 8.07 | — | — | 5.93 | 5.17 | 4.77 | 4.31 | — | — | 3.71 | 3.38 | — | 20 | 5 |
| N ₂ –Ar | 10.93 | 9.57 | — | 8.35 | 7.40 | 6.51 | 6.22 | 5.98 | 5.61 | 5.31 | 4.80 | — | 20 | 5 |
| O ₂ –O ₂ | 11.12 | 9.88 | 9.53 | 8.69 | 7.60 | 6.52 | 6.22 | 5.99 | 5.64 | 5.39 | 4.94 | — | 20 | 24 |
| O ₂ –NO | 11.39 | 10.10 | 9.75 | 8.89 | 7.74 | 6.56 | 6.23 | 5.98 | 5.59 | 5.31 | 4.82 | — | 25 | 24 |
| O ₂ –N | — | 7.56 | 7.26 | 6.55 | 5.60 | 4.75 | 4.49 | 4.28 | 3.96 | 3.72 | 3.31 | — | 25 | 16 |
| O ₂ –O | 9.10 | — | 7.58 | 6.74 | 5.70 | 4.78 | — | 4.29 | 3.96 | 3.71 | — | — | 10 | 8 |
| O ₂ –Ar | 11.30 | 9.86 | — | 8.52 | 7.42 | 6.39 | 6.08 | 5.83 | 5.44 | 5.16 | 4.67 | — | 20 | 5 |
| NO–NO | 11.66 | 10.33 | 9.97 | 9.09 | 7.90 | 6.60 | 6.24 | 5.96 | 5.54 | 5.23 | 4.70 | — | 20 | 24 |
| NO–N | — | 8.21 | 7.86 | 6.99 | 5.90 | 4.91 | 4.61 | 4.37 | 4.01 | 3.73 | 3.27 | — | 25 | 16 |
| NO–O | — | 7.57 | 7.27 | 6.55 | 5.62 | 4.78 | 4.52 | 4.31 | 4.00 | 3.76 | 3.35 | — | 25 | 16 |
| NO–Ar | 11.44 | 10.10 | 9.73 | 8.86 | 7.78 | 6.66 | 6.32 | 6.06 | 5.67 | 5.37 | 4.86 | — | 25 | 24 |
| N–N | 8.07 | 7.03 | — | 5.96 | 5.15 | 4.39 | 4.14 | 3.94 | 3.61 | 3.37 | 2.92 | 2.62 | 5 | 15 |
| N–O | 8.32 | 7.34 | — | 6.22 | 5.26 | 4.45 | 4.21 | 4.01 | 3.69 | 3.43 | 2.98 | 2.66 | 5 | 15 |
| N–Ar | — | 7.94 | 7.62 | 6.79 | 5.72 | 4.76 | 4.47 | 4.25 | 3.89 | 3.64 | 3.19 | — | 20 | 16 |
| O–O | 8.53 | 7.28 | — | 5.89 | 4.84 | 4.00 | 3.76 | 3.57 | 3.27 | 3.05 | 2.65 | 2.39 | 5 | 15 |
| O–Ar | 9.62 | 8.72 | — | 7.58 | 6.52 | 5.54 | 5.24 | 5.00 | 4.64 | 4.37 | 3.90 | 3.58 | 20 | 5 |
| Ar–Ar | 11.67 | 10.05 | — | 8.61 | 7.50 | 6.47 | 6.16 | 5.90 | 5.51 | 5.22 | 4.72 | 4.41 | 5 | 24 |
| e–N ₂ | — | 1.56 | — | 2.17 | 2.91 | 3.59 | 3.80 | 3.93 | 3.99 | 3.91 | 3.57 | 3.29 | 25 | 28, 35 |
| e–O ₂ | — | — | — | 1.31 | 1.72 | 1.99 | 2.04 | 2.06 | 2.06 | 2.05 | 1.99 | 1.96 | 20 | 28, 32 |
| e–NO | — | — | — | — | 4.53 | 4.64 | 4.29 | 3.97 | 3.48 | 3.17 | 2.75 | 2.55 | 35 | 28, 37 |
| e–N | — | — | — | — | 9.04 | 4.06 | 3.33 | 2.93 | 2.53 | 2.34 | 2.13 | 1.98 | 35 | 39, 40 |
| e–O | — | — | — | 0.72 | 0.85 | 0.98 | 1.02 | 1.05 | 1.09 | 1.13 | 1.20 | 1.26 | 30 | 27 |
| e–Ar | — | — | — | 0.12 | 0.19 | 0.43 | 0.55 | 0.66 | 0.89 | 1.12 | 1.68 | 2.23 | 15 | 42, 43 |

Table 2 Viscosity collision integral $\Omega^{2,2}$ (\AA^2) as a function of temperature for neutral–neutral and electron–neutral interactions in air

| Interaction | T, K | | | | | | | | | | | | Acc.,% | Ref. |
|--------------------------------|-------|-------|-------|-------|------|------|------|------|------|--------|--------|--------|--------|--------|
| | 300 | 500 | 600 | 1000 | 2000 | 4000 | 5000 | 6000 | 8000 | 10,000 | 15,000 | 20,000 | | |
| N ₂ –N ₂ | 13.72 | — | 11.80 | 10.94 | 9.82 | 8.70 | — | 8.08 | 7.58 | 7.32 | — | — | 10 | 9 |
| N ₂ –O ₂ | 11.23 | — | — | 8.36 | 7.35 | 6.47 | 6.21 | — | — | 5.42 | 4.94 | — | 20 | 5 |
| N ₂ –NO | 13.44 | 11.87 | 11.44 | 10.48 | 9.32 | 8.04 | 7.61 | 7.27 | 6.74 | 6.33 | 5.62 | — | 25 | 24 |
| N ₂ –N | 11.21 | — | 9.68 | 8.81 | 7.76 | 6.73 | — | 6.18 | 5.74 | 5.36 | — | — | 10 | 8 |
| N ₂ –O | 8.99 | — | — | 6.72 | 5.91 | 5.22 | 5.01 | — | — | 4.36 | 3.95 | — | 20 | 5 |
| N ₂ –Ar | 11.97 | 10.50 | — | 9.28 | 8.38 | 7.53 | 7.25 | 7.02 | 6.63 | 6.32 | 5.73 | — | 20 | 5 |
| O ₂ –O ₂ | 12.62 | 11.06 | 10.65 | 9.72 | 8.70 | 7.70 | 7.38 | 7.12 | 6.73 | 6.42 | 5.89 | — | 20 | 24 |
| O ₂ –NO | 12.93 | 11.32 | 10.90 | 9.94 | 8.89 | 7.80 | 7.45 | 7.17 | 6.73 | 6.39 | 5.80 | — | 25 | 24 |
| O ₂ –N | — | 8.79 | 8.47 | 7.68 | 6.63 | 5.67 | 5.38 | 5.14 | 4.78 | 4.51 | 4.04 | — | 25 | 16 |
| O ₂ –O | 10.13 | — | 8.61 | 7.78 | 6.71 | 5.67 | — | 5.13 | 4.78 | 4.50 | — | — | 10 | 8 |
| O ₂ –Ar | 12.33 | 10.82 | — | 9.57 | 8.54 | 7.48 | 7.14 | 6.87 | 6.44 | 6.12 | 5.54 | — | 20 | 5 |
| NO–NO | 13.25 | 11.58 | 11.15 | 10.16 | 9.07 | 7.91 | 7.53 | 7.21 | 6.73 | 6.36 | 5.72 | — | 20 | 24 |
| NO–N | — | 9.65 | 9.26 | 8.29 | 7.07 | 5.94 | 5.60 | 5.33 | 4.91 | 4.60 | 4.06 | — | 25 | 16 |
| NO–O | — | 8.79 | 8.47 | 7.66 | 6.64 | 5.69 | 5.40 | 5.17 | 4.82 | 4.55 | 4.08 | — | 25 | 16 |
| NO–Ar | 13.03 | 11.33 | 10.89 | 9.90 | 8.85 | 7.79 | 7.44 | 7.15 | 6.71 | 6.37 | 5.77 | — | 25 | 24 |
| N–N | 9.11 | 7.94 | — | 6.72 | 5.82 | 4.98 | 4.70 | 4.48 | 4.14 | 3.88 | 3.43 | 3.11 | 5 | 15 |
| N–O | 9.08 | 8.15 | — | 7.09 | 6.06 | 5.14 | 4.88 | 4.67 | 4.34 | 4.07 | 3.56 | 3.21 | 5 | 15 |
| N–Ar | — | 9.31 | 8.96 | 8.03 | 6.84 | 5.75 | 5.42 | 5.17 | 4.77 | 4.47 | 3.95 | — | 20 | 16 |
| O–O | 9.46 | 8.22 | — | 6.76 | 5.58 | 4.67 | 4.41 | 4.20 | 3.88 | 3.64 | 3.21 | 2.91 | 5 | 15 |
| O–Ar | 11.11 | 10.13 | — | 8.87 | 7.69 | 6.60 | 6.26 | 6.00 | 5.59 | 5.28 | 4.74 | 4.37 | 20 | 5 |
| Ar–Ar | 12.88 | 11.12 | — | 9.66 | 8.60 | 7.57 | 7.23 | 6.96 | 6.53 | 6.20 | 5.60 | 5.17 | 5 | 24 |
| e–N ₂ | — | 1.46 | — | 2.07 | 2.96 | 3.88 | 4.09 | 4.15 | 4.04 | 3.85 | 3.41 | 3.12 | 25 | 28 |
| e–O ₂ | — | — | — | 1.30 | 1.73 | 2.10 | 2.18 | 2.23 | 2.29 | 2.31 | 2.32 | 2.31 | 20 | 28 |
| e–NO | — | — | — | — | 5.64 | 4.52 | 4.05 | 3.73 | 3.37 | 3.18 | 2.92 | 2.75 | 35 | 28 |
| e–N | — | — | — | — | 5.68 | 3.71 | 3.52 | 3.42 | 3.30 | 3.20 | 2.95 | 2.58 | 35 | 39, 40 |
| e–O | — | — | — | 0.82 | 1.05 | 1.34 | 1.44 | 1.52 | 1.65 | 1.73 | 1.85 | 1.90 | 30 | 27 |
| e–Ar | — | — | — | 0.17 | 0.30 | 0.79 | 1.05 | 1.31 | 1.81 | 2.25 | 3.07 | 3.48 | 15 | 26 |

Table 3 Recommended (constant) values for the collision integral ratios B^* and C^* for neutral–neutral and ion–neutral interactions in air

| Interaction type | B^* | C^* | Acc.,% | Ref. |
|------------------|-------|-------|--------|------|
| Neutral–neutral | 1.15 | 0.92 | 10 | — |
| Ion–neutral | 1.20 | 0.85 | 10 | 10 |

temperature range of interest with more accurate ab initio data for the N–O⁺, N–N⁺, O–O⁺, and O–N⁺ interactions.¹⁰

Electron–Neutral Interactions

There is no good engineering approximation to estimate collision integrals for electron–neutral interactions. These data are typically obtained via beam scattering or swarm experiments, the results of which are generally published as integral elastic or momentum

transfer cross sections as a function of energy. Viscosity cross-sectional data are seldom found in the literature for these interactions; therefore many researchers assume that the electron scattering is isotropic,²⁸ which implies that the resulting momentum transfer and viscosity collision integrals are approximately equal ($\Omega^{1.1} = \Omega^{2.2}$).^{4–6} However, this assumption has been shown to be inaccurate^{28,29} for cases where sufficient data are available. Accordingly, Capitelli et al.⁷ use phase shifts to compute the viscosity cross section when only momentum transfer or total elastic cross-sectional data are available.

The most accurate method for determining collision integrals for these interactions is by numerical integration of differential elastic cross sections (DCS), if available. These differential cross sections ($d\sigma/d\Omega$) can be numerically integrated over all scattering angles to obtain integral momentum transfer (diffusion) and viscosity cross sections as a function of the interaction energy¹:

$$\sigma_m(E) = 2\pi \int_0^\pi \frac{d\sigma}{d\Omega} \sin\theta (1 - \cos\theta) d\theta \quad (1)$$

$$\sigma_v(E) = 2\pi \int_0^\pi \frac{d\sigma}{d\Omega} (\sin^3\theta) d\theta \quad (2)$$

The resulting cross sections can then be integrated again over energy assuming a Boltzmann distribution¹⁷ to obtain the necessary transport collision integrals as a function of temperature. The sources used for the necessary cross-sectional data for each of the six electron-neutral interactions in air are discussed later.

Because the collision integrals result from integrations of the differential cross sections over both angular distribution and interaction energy, the resulting values are not sensitive to details of the fine structure (e.g., near resonances) of the DCS. In addition, because both σ_m and σ_v include $\sin\theta$ in the integral, they are not sensitive to values of the DCS near scattering angles of 0 and 180 deg (Ref. 30). [The sensitivity of σ_m to high-angle scattering will be much higher than for σ_v because of the $1 - \cos\theta$ term in Eq. (1).] However, systematic uncertainties in the input data (e.g., all DCS overpredicted) are linearly propagated to uncertainty in the resulting collision integrals. Systematic uncertainties can be a particular concern for experimental data, where the determination of absolute (as opposed to relative) DCS generally involves normalization by a reference cross section.³¹ Uncertainty estimates for each of the six electron-neutral interactions discussed are based on published values in the cited references when available.

For the e–O₂ interaction we follow the recommendation from the recent critical review of electron–molecule interactions by Brunger and Buckman.³⁰ For DCS above 1 eV we use the data of Sullivan et al.³² and Green et al.,³³ which are in good agreement (~10%) with each other, although an estimate of their absolute error is not given. These data are supplemented at lower energies with integral cross-sectional data from Itikawa et al.³⁴ and Phelps,³⁵ which have a stated uncertainty of 20%. The resulting collision integrals are in excellent agreement (~5–10%) with those of Murphy and Arundell⁵ and Capitelli et al.,⁷ which were based exclusively on the Phelps cross-sectional data.³⁵ On the basis of these numbers we estimate a total uncertainty of $\pm 20\%$ in the resulting collision integrals.

For the e–N₂ interaction we again follow the recommendation of Brunger and Buckman.³⁰ DCS data between 0.55 and 10 eV were taken from Sun et al.³⁶ whereas those above 10 eV were taken from Nickel et al.³⁷ At lower energies the integral momentum transfer cross sections recommended by Itikawa³⁸ are employed. The accuracy of these data is not stated, but the scatter between the various experimental results is less than 15% over most of the energy range. Larger discrepancies are observed near 1.8 eV because of the 2\P_g resonance,^{30,38} but as discussed previously these fine structure discrepancies do not have a significant effect on the resulting collision integrals. Therefore, we estimate a total uncertainty of $\pm 25\%$ for this interaction. The prior reviews of Refs. 5–7 all relied on momentum transfer cross sections reported by Phelps and Pitchford.³⁹ These data are generally lower than the current results, with differences ranging from 15% at 2000 K to about 35% at 10,000 K.

These differences are consistent with the estimated uncertainty of the Phelps and Pitchford data.³⁹

Very little experimental data exist for the e–NO interaction, and theoretical calculations are difficult because NO is an open-shell system with a permanent dipole moment.³⁰ DCS data above 1.5 eV are taken from Mojarrabi et al.,⁴⁰ again following the recommendation of Brunger and Buckman.³⁰ The estimated uncertainty in the DCS calculations by Mojarrabi et al. ranges from 5 to 15% (Ref. 30). However, the uncertainty in the resulting integral cross sections was given as 20%, primarily because the DCS data were extrapolated at both low and high scattering angles. At lower energies we use integral momentum transfer cross sections tabulated by Phelps,³⁵ scaled to blend with the more recent higher-energy data of Mojarrabi et al.⁴⁰ The uncertainty in the Phelps data is not given, but the data agree reasonably well with swarm experiments.³⁵ We estimate the total uncertainty for this interaction to be $\pm 35\%$. Collision integrals from Murphy⁶ were based exclusively on the Phelps data³⁵ and are 35–50% higher than the current results.

For the e–O interaction, Itikawa and Ichimura⁴¹ have published a recent review of available cross-sectional data. According to this review, the only available data for transport property calculations are from Thomas and Nesbet,²⁹ who computed integral momentum transfer and viscosity cross sections from their own DCS data.⁴² No uncertainty estimates are given. However, integral elastic cross sections computed by Blaha and Davis⁴³ are in good agreement with those computed by Thomas and Nesbet⁴² and have an estimated uncertainty of 15% above 10 eV (Ref. 43). The uncertainty at lower energies was expected to be larger but was not stated. Given the lack of quantitative data on this interaction, we estimate a total uncertainty of $\pm 30\%$ in the resulting collision integrals. The $\Omega^{1.1}$ collision integrals resulting from integration of these data (Table 1) are in excellent agreement with those of Murphy and Arundell⁵ and Capitelli et al.,⁷ who used the same DCS in their reviews.

The only available DCS for the e–N interaction are those computed by Thomas and Nesbet⁴⁴ at energies below 11 eV, and the results of Blaha and Davis at energies from 1 to 500 eV (Ref. 43). Blaha and Davis give an estimated uncertainty of 15% in their computed total elastic cross section for energies above 10 eV (Ref. 43). No uncertainty estimates are given for lower energies, but the total elastic cross sections from these two sources agree to within 20% in the overlap region (1–11 eV). The total elastic cross sections are considerably larger (~100%) than the experimental results of Neynaber et al.⁴⁵ but are in reasonable agreement (<25%) with other experimental data.^{43,44} Previous compilations of this collision integral relied exclusively on the total elastic cross sections of Neynaber et al.,⁴⁵ either unmodified⁵ or scaled with computed phase shifts.⁷ Collision integrals computed using these data are a factor of 2–4 smaller than the current results in the temperature range 5000–20,000 K,^{5,7} which is not surprising because the cross sections of Neynaber et al.⁴⁵ were all above 2 eV and thus did not show the large maximum below 1 eV observed in the computations of Thomas and Nesbet and lower energy experimental results.⁴⁴ Based on accuracy of the data of Refs. 43 and 44, we estimate a total uncertainty of $\pm 35\%$ for this interaction. However, it should be noted that, given the large discrepancies in the various published results, additional experimental or theoretical data for this interaction would be most welcome.

Finally, for the e–Ar interaction, no low-energy DCS data are available. However, integral momentum transfer cross sections are taken from Milloy et al.⁴⁶ over the energy range 0–4 eV and Frost and Phelps⁴⁷ over the energy range 4–20 eV, following the recommendation of Murphy and Arundell.⁵ The stated uncertainty of the Milloy et al. data is $\pm 8\%$ except in the energy range 0.1–0.4 eV, where the so-called Ramsauer minimum occurs and errors are estimated at -20% to $+12\%$ (Ref. 46). Frost and Phelps estimated their uncertainty as about $\pm 10\%$ above 1 eV (Ref. 47). The agreement between the Frost and Phelps and the Milloy et al. data is quite good (<10%) in the overlap region 1–4 eV. We therefore estimate the total uncertainty in the momentum transfer cross sections at about $\pm 15\%$ over the entire energy range. Integral viscosity cross sections are taken from Zigman and Milic,²⁸ who estimate a total uncertainty of less than $\pm 12\%$ over the energy range 0.14–15 eV.

Table 4 Collision integral ratio B^* as a function of temperature for electron-neutral interactions in air

| Interaction | T, K | | | | | | | | | | | | Acc.,% | Ref. |
|------------------|------|------|-----|------|-------|-------|-------|-------|-------|--------|--------|--------|--------|--------|
| | 300 | 500 | 600 | 1000 | 2000 | 4000 | 5000 | 6000 | 8000 | 10,000 | 15,000 | 20,000 | | |
| e-N ₂ | — | 0.69 | — | 0.61 | 0.82 | 0.83 | 0.96 | 1.08 | 1.22 | 1.25 | 1.20 | 1.14 | 20 | 28, 36 |
| e-O ₂ | — | — | — | 0.68 | 0.86 | 0.99 | 1.02 | 1.04 | 1.07 | 1.07 | 1.03 | 1.00 | 20 | 28, 32 |
| e-NO | — | — | — | — | 1.09 | 1.38 | 1.34 | 1.28 | 1.19 | 1.13 | 1.07 | 1.07 | 20 | 28, 33 |
| e-N | — | — | — | — | 1.52 | 1.04 | 1.00 | 0.99 | 1.01 | 1.03 | 1.16 | 1.40 | 20 | 39, 40 |
| e-O | — | — | — | 0.81 | 0.85 | 0.89 | 0.90 | 0.91 | 0.90 | 0.89 | 0.87 | 0.86 | 20 | 27 |
| e-Ar | — | — | — | — | -0.40 | -0.05 | -0.02 | -0.01 | -0.02 | -0.04 | 0.03 | 0.29 | 15 | 42, 43 |

Table 5 Collision integral ratio C^* as a function of temperature for electron-neutral interactions in air

| Interaction | T, K | | | | | | | | | | | | Acc.,% | Ref. |
|------------------|------|------|-----|------|------|------|------|------|------|--------|--------|--------|--------|--------|
| | 300 | 500 | 600 | 1000 | 2000 | 4000 | 5000 | 6000 | 8000 | 10,000 | 15,000 | 20,000 | | |
| e-N ₂ | — | 1.18 | — | 1.16 | 1.12 | 1.09 | 1.08 | 1.05 | 0.99 | 0.95 | 0.91 | 0.91 | 20 | 28, 36 |
| e-O ₂ | — | — | — | 1.16 | 1.10 | 1.04 | 1.03 | 1.01 | 0.99 | 0.98 | 0.98 | 0.98 | 20 | 28, 32 |
| e-NO | — | — | — | — | 1.15 | 0.90 | 0.87 | 0.85 | 0.85 | 0.87 | 0.90 | 0.93 | 20 | 28, 33 |
| e-N | — | — | — | — | 0.63 | 0.67 | 0.74 | 0.79 | 0.87 | 0.90 | 0.93 | 0.89 | 20 | 39, 40 |
| e-O | — | — | — | 1.09 | 1.08 | 1.06 | 1.05 | 1.05 | 1.05 | 1.05 | 1.05 | 1.06 | 20 | 27 |
| e-Ar | — | — | — | — | 1.40 | 1.36 | 1.35 | 1.34 | 1.34 | 1.34 | 1.34 | 1.31 | 15 | 42, 43 |

Recommended values of $\Omega^{1,1}$ and $\Omega^{2,2}$ for the six electron-neutral interactions based on these references are given in Tables 1 and 2. In most cases values are not given below 1000 K because of a lack of cross-sectional data at low interaction energies. However, the relative importance of electron-neutral interactions at very low temperatures should be negligible for most applications. The collision integral ratios B^* and C^* derived from these data show more temperature dependence as well as variability between the different electron-neutral interactions than was observed previously for neutral-neutral and ion-neutral interactions. Therefore, B^* and C^* are given for each of the six interactions as a function of temperature in Tables 4 and 5.

Charged-Particle Interactions

Finally, the 28 interactions between charged particles (ion-ion, ion-electron, and electron-electron) must be considered. Fortunately, Mason et al.⁴⁸ have shown that under most conditions these interactions can be adequately represented using an attractive (ion-electron) or repulsive (ion-ion and electron-electron) shielded coulomb potential given by

$$\varphi(r) = \pm [e^2/r]e^{-r/\lambda_D} \quad (3)$$

In Eq. (3) the sign indicates an attractive (+) or a repulsive (−) potential, and λ_D is the Debye shielding length in centimeters, given by

$$\lambda_D = \sqrt{\frac{kT}{4\pi n_e e^2}} \quad (4)$$

The classical transport collision integrals result from integration of this potential¹ and are functions of only of the dimensionless reduced temperature, given by

$$T^* = \lambda_D/(e^2/kT) \quad (5)$$

which is itself a function only of the temperature and the electron-number density. Mason et al.⁴⁸ have published tables of the resulting collision integrals for attractive and repulsive shielded coulomb potentials. The resulting diffusion and viscosity collision integrals can be fit to an expression of the form⁴⁹

$$\Omega^{n,n} = 5.0 \times 10^{15} (\lambda_D/T^*)^2 \ell_n \{ D_n T^* [1 - C_n \exp(-c_n T^*)] + 1 \} \quad (6)$$

Values of the coefficients D_n , C_n , and c_n for computing $\Omega^{1,1}$ and $\Omega^{2,2}$ for both attractive and repulsive potentials are given in Table 6, reproduced from Ref. 2.

Table 6 Curve-fit coefficients for the classical shielded coulomb potential

| Collision integral | Attractive potential | | | Repulsive potential | | |
|--------------------|----------------------|--------|-------|---------------------|--------|-------|
| | C_n | c_n | D_n | C_n | c_n | D_n |
| $\Omega^{1,1}$ | -0.476 | 0.0313 | 0.784 | 0.138 | 0.0106 | 0.765 |
| $\Omega^{2,2}$ | -0.146 | 0.0377 | 1.262 | 0.157 | 0.0274 | 1.235 |

Table 7 Curve-fit coefficients for the classical shielded coulomb potential

| Ratio | Attractive potential | | | Repulsive potential | | |
|-------|----------------------|--------|---------|---------------------|--------|---------|
| | a | b | c | a | b | c |
| B^* | 1.0355 | 0.2864 | -4.4229 | 1.0322 | 0.4116 | -3.9448 |
| C^* | 0.3538 | 0.1642 | -3.8017 | 0.3531 | 0.2587 | -3.1829 |

Equation (6) cannot be used to fit collision integrals for $l \neq n$ (e.g., $\Omega^{1,2}$ and $\Omega^{1,3}$), because the asymptotic form is incorrect for large T^* . However, the dimensionless collision integral ratios B^* and C^* can be readily fit to a Gaussian expression of the form

$$(B^*, C^*) = a + b \exp[\ell_n(T^*)/c] \quad (7)$$

Values of the coefficients a , b , and c in Eq. (7) are generated for this work for both attractive and repulsive potentials and are presented in Table 7. It should be noted that the constants in Tables 6 and 7 are strictly valid for only $T^* > 4$. However, this range covers the temperatures and pressures of interest for most reentry and laboratory plasmas. As an example, for an equilibrium air mixture at 100 kPa, this condition is satisfied for all temperatures.

At low temperatures and/or high electron-number densities, the shielded Coulomb collision integrals may need to be corrected for quantum-mechanical effects.^{49,50} This effect would be most pronounced for electron-electron interactions, because of the small reduced mass of the system. Hahn et al.⁵⁰ have shown that a reasonable approximation to the necessary quantum corrections can be derived by assuming that at close range the interaction is governed by a simple Coulomb interaction potential. This approximation results in correction terms to the classical shielded Coulomb values. The final collision integrals are then given by

$$\Omega^{l,n}(T^*) = \Omega_{\text{class}}^{l,n}(T^*) - \Omega_{\text{qm}}^{l,n}(T^* \Lambda^{*2}) \mp [1/(2s+1)] \Omega_{\text{ex}}^{l,n}(T^* \Lambda^{*2}) \quad (8)$$

where the de Boer parameter Λ^* is defined by

$$\Lambda^{*2} = h^2/2\mu e^2 \lambda_D \quad (9)$$

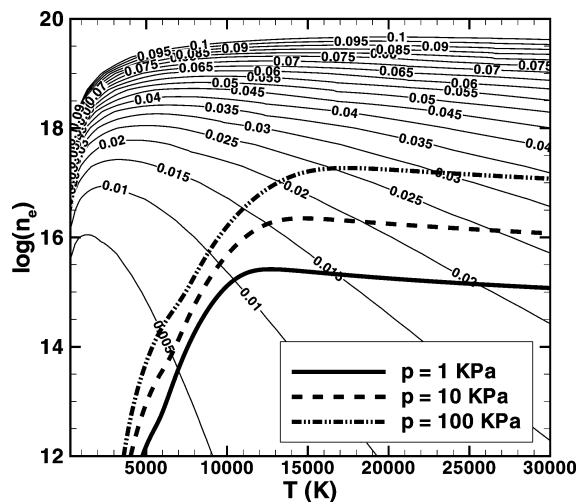


Fig. 3 Contours of the ratio of the sum of the two quantum mechanical correction terms from Ref. 50 to the classical (shielded coulomb) viscosity collision integral ($\Omega^{2,2}$) for e-e interactions, overlaid with lines showing the electron-number density of equilibrium air as a function of temperature for three pressures.

As shown in Eq. (8), two correction terms are derived in Ref. 50, one due to quantum mechanical effects ($\Omega_{qm}^{l,n}$) and an exchange term ($\Omega_{ex}^{l,n}$) that is nonzero only for interactions between indistinguishable particles. Note that quantum indistinguishability is only meaningful in interactions with even values of l (Ref. 50), and thus $\Omega_{ex}^{l,n}$ should be set to zero in Eq. (8) for l odd. The sign on the exchange term in Eq. (8) results from the use of either Fermi–Dirac (–) or Bose–Einstein (+) statistics. For electron–electron interactions $s = \frac{1}{2}$ and Fermi–Dirac statistics apply. The resulting correction terms are tabulated in Ref. 50.

Figure 3 shows the ratio of the combined quantum-mechanical and exchange contributions to the viscosity collision integral divided by the classical collision integral for electron–electron interactions as a function of temperature and electron-number density. The classical shielded coulomb potential was computed by using Eq. (6) for $T^* > 4$, and the corresponding expression from Ref. 49 for lower values of T^* . Also shown on the figure are lines indicating equilibrium air electron-number density at a constant pressure of 1, 10, and 100 kPa. It is evident from Fig. 3 that for most aerospace and laboratory applications the quantum-mechanical and exchange contributions to the viscosity collision integral are small, ranging from <0.5 to $\sim 3\%$ of the classical value. The quantum-mechanical and exchange terms become significant only at very high levels of electron-number density and/or very low temperatures. The importance of the quantum-mechanical terms in electron–ion interactions will be much smaller because the de Boer parameter is inversely proportional to the reduced mass of the system. Therefore, for most aerospace and laboratory applications quantum-mechanical corrections to the shielded coulomb potential can be safely neglected.

These results assume that the charged particles are statically shielded. However, dynamic shielding effects, which arise from a time lag in the response to particle motion by the surrounding bath of charged particles, can become important under some conditions, primarily at low temperatures and high electron-number densities. The basic theory of dynamic shielding was discussed by Williams and DeWitt⁵¹ and later Hahn et al.,⁵² who showed that dynamic shielding effects changed the thermal conductivity of atmospheric equilibrium Ar plasmas by a maximum of about 10% for a fully ionized gas. Later, Monchick and Mason⁵³ explored the effects of dynamic shielding on both ordinary and thermal diffusion in ionized hydrogen plasmas. The results of Monchick and Mason indicate that dynamic shielding can have a significant effect for dense, highly ionized plasmas, such as would be encountered during Jupiter entry, but is unlikely to be a large contributor to the transport properties of Earth reentry plasmas.

The accuracy of the collision integrals computed using the classical shielded Coulomb potential can be estimated by comparison to the results of Stallcop et al.,⁴⁹ who performed ab initio quantum-mechanical calculations of the transport collision integrals for N^+-N^+ , N^+-O^+ , and O^+-O^+ interactions based on realistic interaction potentials. Stallcop et al. showed⁴⁹ that the classical shielded Coulomb result was within 15% of the more accurate computations for temperatures below 50,000 K, except at very high pressures (small Debye radii).

Conclusions

The data presented in this paper, coupled with the ion-neutral data from Refs. 10 and 11, form a complete set of collision integrals for the computation of the transport properties of 13-species weakly ionized air. Although the fidelity of the data varies, all collision integrals presented herein, with the exception of the electron-neutral interactions, are estimated to be accurate to within 25% over the temperature range of interest (~ 300 – $15,000$ K). In addition, most of the dominant interactions for weakly ionized air have been computed via ab initio methods that are estimated to be accurate to within 10%.

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